

Nonequilibrium Polaritonics - Nonlinear Effects and Optical Switching

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We report a theoretical non-equilibrium description of polaritonics and we propose ultrafast all-optical switching due to highly nonlinear polaritonics. The electronic band structure within gold (Au) nano grains is modified by external laser light. The Au grains are coupled to a single mode photonic waveguide and we derive a strong transmission reduction of switching originating from the established quantum interference with a finite lifetime.

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INTRODUCTION

Plasmonics and polaritonics [1] may be the most promising candidates in the race of novel ultrafast technologies which have the potential to reach the application border [2–4]. Meta-materials or plasmonic systems [7] are on the border to be engineered for ultrafast electronic switches [9–14], and ultra-short laser science opens the door to their exploration [15–17]. However discussing picosecond, femto or attosecond time scales the framework of equilibrium physics reaches its limits. When we talk about ultrafast nonlinear processes in commercially fabricated metallic or organic structures, impurities naturally play a key role for the efficiency and the proper operation of such devices. They may either disturb the process or may play the key role of access to new technologies. Another fascinating track of physics in the context of ultrafast switches are beyond doubt quantum-optical functional elements [5]. Yet the combination of both, highly nonlinear nonequilibrium physics and quantum optics is an ansatz which has not been felt out in a broad range up to now. Solid-state-based elements, provide a number of advantages such as scalability, tunable light-matter interactions and comparably easy handling. Besides its immense technological importance, the theoretical description of externally driven quantum systems is a challenge itself, and vividly discussed [18–20]. While the basic underlying equilibrium physics for a system with discrete levels coupled to a continuum of states is well established [21, 22], the profound description of light-matter interaction far away from thermodynamical equilibrium is an active and fascinating area of research.

FRANZ-KELDYSH EFFECT AND FANO RESONANCE

In this article the electronic properties of gold (Au) nano-grains exposed to an external field are investigated and they are coupled to a single mode photonic silicon-on-insulator hollow core waveguide (SOI). In this non-equilibrium system, the external time-periodic field generates photo-induced electronic side-bands which can be attributed to the Franz - Keldysh effect, the AC analogon to the well known Wannier-

Stark effect. Intraband transitions occurring in every metal within the conduction band feature a more or less small absorption rate, whereas the governing processes are the inter-band transitions. In Au a significantly different behavior has been found, which has been attributed to the electronic specific structure of closed packed Au, namely to the high polarizability of the $5d^{10}$ cores. The collective resonance exhibits a large red shift to approximately 2.4eV which leads to the fact that the corresponding intensity is governed by the interband processes but its existence results from the occurrence of intraband transitions which can be recognized as step like structures in the spectrum. These transitions, which result from a non fermi alike distribution of states, have been observed by Whetten *et al.*[6], for Au nano-spheres of diameters below 30nm . Their occurrence has been interpreted as the border from bulk like characteristics to the quantum regime of nano particles. The transitions within this single plasmon band provide the basis for the switching effect. We show that the band-structure of the coupled system of Au nano-grains and SOI is strongly modified by intense external laser radiation (Fig.4), which can be tested experimentally e.g. by two photon photoemission (2PPE). Frequency and amplitude of the external field can be separately used to modulate the position in energy of the Floquet-side-bands and, therefore sensitively control the generation of a Fano-resonance with the photonic SOI mode. The calculated lifetimes of the coupled states prove the extremely fast switching.

FRÖHLICH HAMILTONIAN AND FLOQUET-KELDYSH METHOD

As theoretical setup the Fröhlich Hamiltonian [23, 24] for fermion-boson interaction is chosen which is solved by applying the Keldysh formalism with respect to the non-equilibrium character of the considered processes on the femto-second time scale. The full Hamiltonian reads

$$H = \sum_{k,\sigma} \epsilon_k c_{k,\sigma}^\dagger c_{k,\sigma} + \hbar\omega_o a^\dagger a + g \sum_{k,\sigma} c_{k,\sigma}^\dagger c_{k,\sigma} (a^\dagger + a) \quad (1)$$

$$- t \sum_{\langle ij \rangle, \sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + i \vec{d} \cdot \vec{E}_0 \cos(\Omega_L \tau) \sum_{\langle ij \rangle} (c_{i,\sigma}^\dagger c_{j,\sigma} - c_{j,\sigma}^\dagger c_{i,\sigma})$$

This article is organized as follows, we consider (i) the waveguide in contact with nano-grains, (ii) the nano-grain in the external field and finally (iii) discuss the non-equilibrium solution of the complete system in terms of electronic Keldysh - Green's function and waveguide transmission. The overall switching is measured as the difference between density of electronic states of the totally unaffected system compared to the setup including all effects due to the external laser light and as a consequence the coupling to the waveguide mode.

We start with the single electron band with nearest-neighbor hopping, characterized by the hopping amplitude t , with the dispersion for a cubic lattice for the ion cores $\epsilon_k = 2t \sum_i \cos(k_i a)$, a is the lattice constant and k_i are the components of the wave-vector. We assume a SOI waveguide supporting a single mode $\hbar\omega_0 = 2.34 eV$. The waveguide itself is coated and therefore not exposed to the external laser field. The electrons couple with strength g weakly to waveguide photons with frequency ω_0 . A possible setup is depicted in Fig.1. The Hamiltonian without the external laser field reads,

$$H = \sum_{k,\sigma} \epsilon_k c_{k,\sigma}^\dagger c_{k,\sigma} + \hbar\omega_0 a^\dagger a + g \sum_{k,\sigma} c_{k,\sigma}^\dagger c_{k,\sigma} (a^\dagger + a). \quad (3)$$

We assume the spatial extension of the Au nano-grains to be smaller than $30nm$, and consequentially small compared to the wavelength of the photonic mode inside the waveguide. Therefore, the momentum of the photons is much smaller than the electron's momentum and we can set $q_{\text{photon}} \simeq 0$ whenever we consider the electronic subsystem. Thus, a^\dagger (a) does not carry an index in the Hamiltonian. In Eq. (3), ϵ_k is the electronic band energy, $c_{k,\sigma}^\dagger$ ($c_{k,\sigma}$) creates (annihilates) an electron with momentum k and spin σ . $\hbar\omega_0 a^\dagger a$ is the photon energy eigen-state, where a^\dagger (a) creates (annihilates) a photon inside the waveguide with energy $\hbar\omega_0$. The last (coupling) term on the r.h.s. is the standard coupling term between the electronic and the photonic subspaces [25]. Due to the weak interaction of waveguide photons and electrons inside the nano-grains we treat this interaction perturbatively. In second order a self-energy contribution is obtained from Eq. (3). The coupling of the electronic system with a continuous energy spectrum to the photons with a discrete one leads to a so-called Fano resonance [21]. This is observed in the electronic density of states (see Fig.3). Here we show the electron's spectral function for various frequencies of the waveguide mode for coupling strength $(g/t)^2 = 0.09$ at zero temperature for a spectral width $\tau = 0.005$ of the waveguide mode (measured in units of the hopping t) at half filling, yielding a suppression of the spectral function around the Fermi-level (half width τ) where electrons are transferred to the high (low) energy tails of the spectral function. We note, that if the energy of the waveguide mode $\hbar\omega_0$ is distinctly different from the energy $\hbar\omega$ of the band-electrons, the electronic density of states remains approximately unchanged.

Now we discuss the subsystem of a nano-grain exposed to a semiclassical electromagnetic laser field. This is described by the Hamiltonian (Lb = laser + band-electrons)

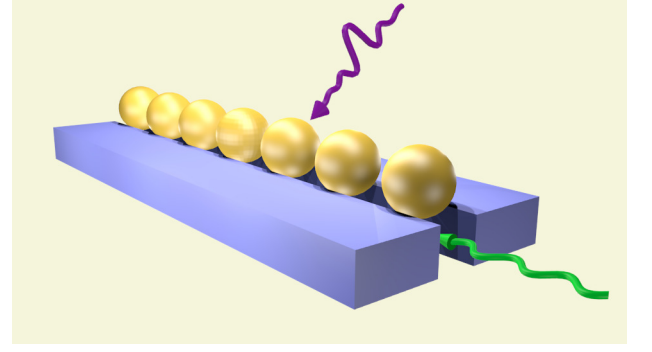


FIG. 1: Sketched geometry. Gold nanograins in contact with a hollow core SOI waveguide. Waveguide photons (green) interact with electrons in the metal nano-grains (Au) forming a coupled light-matter state, a polariton. This is controlled by an external laser (pink), altering the transmission through the waveguide (green).

$$H_{Lb} = -t \sum_{\langle ij \rangle, \sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + H_C(\tau) \quad (4)$$

where $\langle ij \rangle$ implies summation over nearest neighbors. $H_C(\tau)$ represents the coupling to the external, time-dependent laser field, described by the electric field $\vec{E} = \vec{E}_0 \cos(\Omega_L \tau)$, via the electronic dipole operator $\hat{d}(\vec{x})$ with strength \vec{d} . It is given by

$$H_C(\tau) = i \vec{d} \cdot \vec{E}_0 \cos(\Omega_L \tau) \sum_{\langle ij \rangle} \left(c_{i,\sigma}^\dagger c_{j,\sigma} - c_{j,\sigma}^\dagger c_{i,\sigma} \right) \quad (5)$$

The Hamiltonian specifically describes the excitation of a plasmon-polariton, which correspond to spatially delocalized intraband electronic motion caused by an external electromagnetic wave. The accelerating energy is immediately transferred into the motion of electrons by means of single-band nearest neighbor hopping without interaction between the electrons. Due to the time dependence of the external field, Green's functions truly depend on two separate time arguments. Therefore, we use a double Fourier transform from time- to frequency space introducing relative and center-of-mass frequency [26]

$$\begin{aligned} G_{mn}^{\alpha\beta}(\omega) &= \int d\tau_1^\alpha \int d\tau_2^\beta e^{-i\Omega_L(m\tau_1^\alpha - n\tau_2^\beta)} e^{i\omega(\tau_1^\alpha - \tau_2^\beta)} G(\tau_1^\alpha, \tau_2^\beta) \\ &\equiv G^{\alpha\beta}(\omega - m\Omega_L, \omega - n\Omega_L), \end{aligned} \quad (6)$$

where (m, n) label the Floquet modes [27] and (α, β) specify on which branch of the Keldysh contour (\pm) the respective time argument resides. Floquet states are the Fourier-transformed analog to Bloch states: The first ones result from a time-periodic potential whereas the latter are the result of a space-periodic potential and both induce a band-structure. The special case of non-interacting electrons allows an analytical solution for $G_{mn}(k, \omega)$ by solving the equation of motion.

Including photo-induced hopping, the exact retarded Green's function for this sub-system has also been discussed in different context e.g. [20]

$$G_{mn}^R(k, \omega) = \sum_{\rho} \frac{J_{\rho-m}(A_0 \tilde{\epsilon}_k) J_{\rho-n}(A_0 \tilde{\epsilon}_k)}{\omega - \rho \Omega_L - \epsilon_k + i0^+} \quad (7)$$

where $\tilde{\epsilon}_k$ represents the dispersion relation induced by the external field Eq. (5) and is different from ϵ Eq. (3). The J_n are the cylindrical Bessel functions of integer order, $A_0 = \vec{d} \cdot \vec{E}_0$ and Ω_L is the laser frequency. The Bessel function indicate the highly nonlinear characteristics of the switching effect under investigation. The physical Green's function is given according to

$$G_{\text{Lb}}^R(k, \omega) = \sum_{m,n} G_{mn}^R(k, \omega). \quad (8)$$

We present a numerical evaluation of Eq. (8) in Fig.2, where $\text{Im} G_{\text{Lb}}^R(k, \omega)$, is displayed as a function of quasiparticle energy $\hbar\omega$ and external frequency Ω_L at zero temperature for $A_0/t = 2.5$. As a typical value for the hopping we chose $t = 1\text{eV}$. The waveguide is operated at the frequency $\hbar\omega_0 = 2.34\text{eV}$ which corresponds to a frequency doubled Nd-YAG laser ($\hbar\omega_0 = 1.17\text{eV}$). The external laser shall be characterized by 10fs pulses, and shall be $E_0 = 1.008 \times 10^{10}\text{V/m}$ in the surface region of the Au grains, including Mie type [28] field enhancement effects due to the small particle sizes [29–31]. For the nano-grains we choose a damage threshold [32] of 0.5J/cm^2 and $|d| = 6.528 \times 10^{-29}\text{Asm}$ (with the lattice constant for Au $a_{\text{Au}} = 4.08 \times 10^{-10}\text{m}$) resulting [33] in $A_0 \equiv d \cdot E_0 = 4.0\text{eV}$. We assume further a density of 50% nanograins per $10\text{ }\mu\text{m}$ of the waveguide. The original semi-circular density of states develops photonic side-bands, the bandstructure, as the external laser frequency Ω_L increases. Due to the point-inversion symmetry of the underlying lattice, the first side-band represents here the two-photon processes. The less pronounced second side-band, therefore, represents four-photon processes. Their occupation is described by the non-equilibrium distribution function as calculated from the Keldysh component of the Green's function.

In a last step, we combine the relaxation processes due to the interaction between the band electrons and the waveguide as described by Eq. (3), with the impinging external laser as introduced in Eq. (4). The resulting Green's function consequently describes the photonic waveguide with Au nano-grains that themselves are now exposed to the external laser radiation. In this non-equilibrium system we treat the weak coupling between the electrons and the waveguide photons by second order perturbation theory and the interaction between the electrons and external laser in terms of the Floquet theory, as has been shown above.

Since we are interested in possible switching effects, we choose as the initial situation the case where the photonic mode, $\hbar\omega_0 = 2.34\text{eV}$, is far off the electronic bandedge. Electrons arrive band energies ranging from $-1 \leq \hbar\omega/t \leq +1$. A Fano resonance is only observable in the presence of

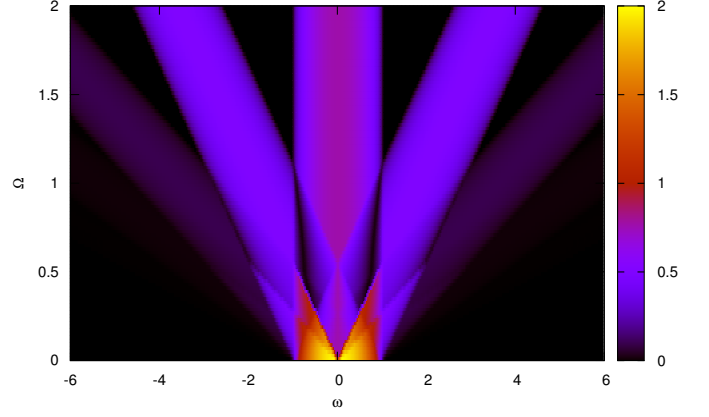


FIG. 2: The imaginary part of the local Green's function, the local density of states (LDOS) Eq. (8), is depicted as a function of quasiparticle energy, and frequency respectively ω and external laser frequency Ω_L at zero temperature for external field amplitude $A_0/t = 4.0\text{ eV}$ (see caption text). The original semicircular DOS evolves photonic side-bands as the laser frequency increases. Side-bands of first and second order can be identified for this specific value of field amplitude.

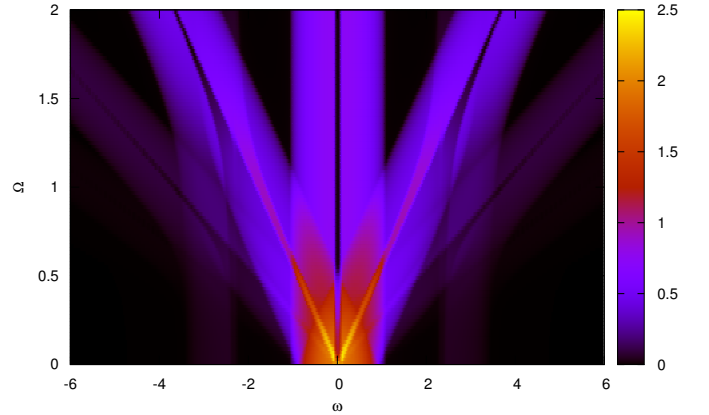


FIG. 3: Depicted is the imaginary part of the full Green's function, including all interactions as a function of frequency ω and external frequency Ω_L at zero temperature. The external amplitude is chosen to be $A_0/t = 4.0$ (see text). The electronic Floquet-Keldysh (Fig. 2) bandstructure is modified by the coupling to the single mode waveguide operated at $\hbar\omega = 2.34\text{ eV}$. The coupling is established by an electron-mediated Fano resonance. This Fano resonance results in an avoided crossing of bands at $\hbar\omega = 2.34\text{ eV}$ and $\hbar\Omega = 1.17\text{ eV}$. Spectral weight from the Fermi edge position in equilibrium is obviously shifted to the center of the Floquet sidebands by that coupling of gold and waveguide which results in the development of a gap in the center of the original band at $\omega = 0.0$.

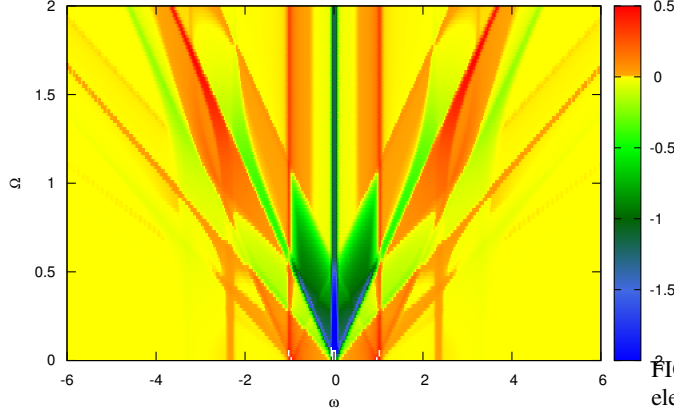


FIG. 4: Laser induced change of the electronic density of states $\delta G(\omega, \Omega_L)$, Eq. (9). The photonic waveguide mode has $\hbar\omega_0 = 2.34\text{eV}$, the coupling between waveguide photons and electrons is $g/t=0.3$ ($\sim 30\%$ of the coupling A_0), the temperature $T = 0$ and the amplitude $A_0/t=4.0$ (see text). δG displays a Fano resonance around quasiparticle frequencies $\omega=\omega_0$, i.e. as soon as the external laser field redistributes the electronic spectral weight such, that the waveguide mode finds electrons with about the same energy to efficiently interact with.

external radiation of appropriate frequency, i.e. only if the induced side-bands meet the energy of the waveguide mode. This resonance will also affect the transmission properties in the waveguide, since now waveguide photons can be absorbed in the formation of a mixed state of light and matter with the laser-induced charge excitations in the Au nano-grains. Thus a waveguide polariton is created yielding to a significant reduction of the waveguide's transmission. In 4, we display the laser induced change of the density of states δG as a function of quasi-particle energy $\hbar\omega$ and external laser frequency Ω_L . The quantity δG defined according to

$$\delta G = [\text{Im } G(\omega, \Omega_L) - \text{Im } G_{\text{Lb}}(\omega, \Omega_L)] - [\text{Im } G_{\text{wb}}(\omega) - \text{Im } G_{\text{b}}(\omega)] \quad (9)$$

measures the effect of the impinging laser field on the electronic density of states, and vanishes as the external laser and the coupling to the waveguide is turned off. In Eq. (9), G represents the Green's function including all processes, G_{Lb} the interaction between the laser field and the band electrons as given in Eq. (8), G_{wb} describes the waveguide in presence of the band electrons and is solution to Eq. (3), and finally G_{b} is the Green's function of just the noninteracting band electrons. In 3 the laser induced electronic density of states coupled to the waveguide $G(\omega, \Omega_L)$ experiences a Fano resonance as soon as the external laser redistributes electronic spectral weight such, that the waveguide mode at $\hbar\omega_0 = 2.34\text{eV}$ may efficiently be absorbed. This is derived when the first photonic side-band meets the energy of the pho-

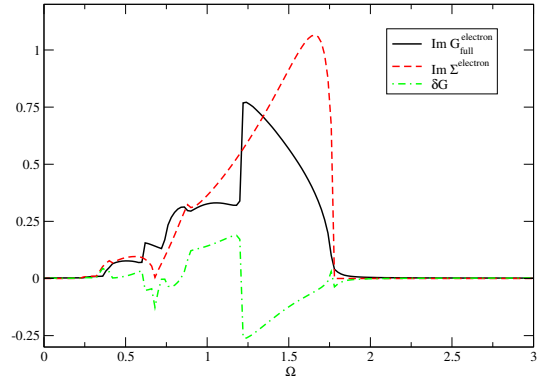


FIG. 5: The imaginary part of the retarded component of the full electronic Green's function $\Im G$ is shown in combination with the imaginary part of the retarded component of the electronic self-energy $\Im \Sigma$ and the imaginary part of the difference Green's function δG as defined in the text, for a single electronic energy of $\hbar\omega = 2.34\text{ eV}$ and for a range of external pump laser frequencies Ω . At a about $\hbar\Omega = 1.2\text{ eV}$ the sharp dip in δG indicates the maximum modulation in the transmittance of the waveguide mode due to the strong coupling to the nanograins. That point of switching corresponds to a maximum change of spectral weight in the electronic density of states $\Im G$. We find the inverse lifetime $\Im \Sigma$ to be finite valued and the corresponding lifetime of the states equals $t = 5.908 \cdot 10^{-15}\text{ s}$. The element can be operated having very short dead times.

tonic waveguide mode yielding a sign change in δG at this energy; compare also to Fig.2. Additionally, we find in Fig.3 at this particular point an avoided crossing of the bands. In Fig.4, the laser induced change of the electronic density of states δG is shown at fixed quasiparticle energy $\hbar\omega = \hbar\omega_0$, where ω_0 is the frequency of the photonic waveguide mode. Asymptotically, i.e., for large Ω_L , δG vanishes, as already indicated in 2, because in this limit there is no electronic spectral weight at the particular energy of the waveguide mode. In the opposite limit, $\Omega_L \rightarrow 0$, the influence of the laser field is non-zero, because here higher-order laser induced side-bands exist, yielding spectral weight at the resonance position already for smaller laser frequencies. That result can also be concluded from the second-order side-band in 3.

In a waveguide of length l , the ratio between the initial and the transmitted intensity is given by $T \sim \exp(-\alpha l/l_o)$. Here α/l_o , is the absorption coefficient divided by the unit length l_o , where α includes an average over one period of the external periodic driving field with frequency Ω_L . We recognize that $\omega\delta G$ can be understood as the leading contribution to the relative absorption coefficient as discussed in detail in ref. [38]. In 8 we show the relative transmission of photons T/T_0 within the waveguide of unit length $l = l_o$ as a function of the external laser frequency Ω_L . Depending on the frequency of the driving field, an intensity drop of up to 25% is observed. By increasing the length of the waveguide or the density of the grains this effect is enhanced. Therefore we conclude, that by switching laser light of an appropriate frequency of about $\hbar\Omega_L/t \sim 1.35$, the transmitted photon intensity through the

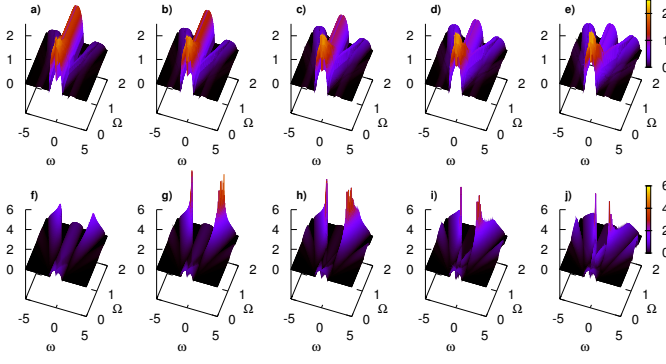


FIG. 6: The upper panel, figures a) through e), displays the imaginary part of the full interacting electronic retarded Green's function component (i.e. up to a factor $1/\pi$ the LDOS) for increasing strengths of the external pump laser. The amplitude A_0 , see text, of the external laser field assumes values of 2.0, 2.5, 3.0, 3.5 and 4.0 from a) to e). An increased weight of the Floquet sidebands with increasing pump strengths is observed, simultaneously the main (zeroth) Floquet band loses weight. Further an increased strength of the Fano resonance in the first Floquet sideband at about $\hbar\omega = 2.34$ eV is found from left to right. The lower panel, graphs f) to j), features the imaginary part of the corresponding retarded component of the electronic selfenergy for the same external amplitudes A_0 assuming 2.0, 2.5, 3.0, 3.5 and 4.0 from f) to j). This imaginary part is proportional to the inverse lifetime of the electronic excitations. From left to right an decrease of lifetimes with increasing pumpstrength is visible. Furthermore, a strong enhancement of the imaginary part of the retarded component of the selfenergy in the sidebands is observed which is moving towards lower pump frequencies Ω when the pump strength is increased, i.e. from left to right in the lower panel.

waveguide can be significantly altered, and by varying the length of the waveguide the transmission inside the waveguide can in fact be turned on and off. The calculated lifetime of the coupled state in the position of the fano resonance for the presented parameters equals $t = 5.908 \cdot 10^{-15}$ s. After that reaction time the switch should be in the initial state and ready for the next signal. In Fig.6 we discuss the evolution of the density of states LDOS and the corresponding lifetimes of these states with increasing external laser amplitude. It is found, that the Floquet-sidebands evolve with raising field amplitude and so does the calculated inverse lifetime. That behavior results actually in a deminishing of the lifetime of coupled states with raising field amplitude and it can be clearly observed, that the minimum of lifetime is shifting with increasing amplitude towards the position of the Fano resonance, which is located at $\omega = 2.34$ eV and $\Omega = 1.17$ eV.

CONCLUSION

In conclusion we have presented a quantum field theoretical model for a photonic waveguide in contact with gold nano-grains which themselves are exposed to external laser irradiation. The strong and coherent external laser is described

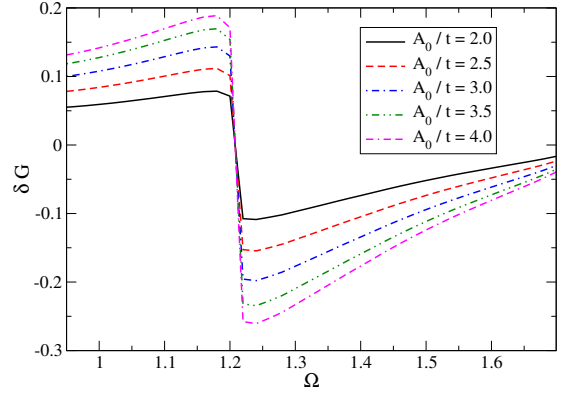


FIG. 7: The relative switch of electronic density of states δG is a measure for the dip in the transmission of waveguide photons and therefore the inverse polaritonic coupling between waveguide photons and electrons. The waveguide is of unit length $l=l_0$. The graph is displayed as a function of external laser frequency Ω_L , for four different amplitudes of the external field. Parameters are given in the text.

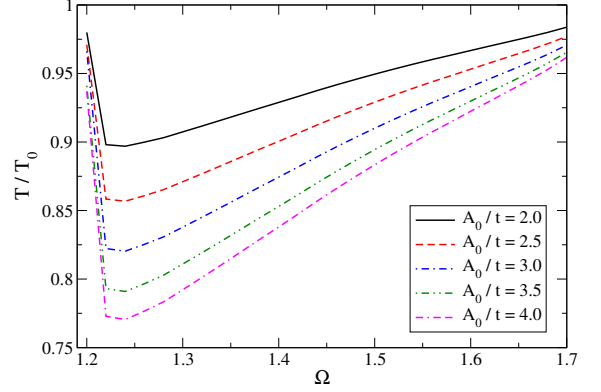


FIG. 8: Relative photon transmission in a waveguide of unit length $l=l_0$ as a function of laser frequency Ω_L , for four different amplitudes of the external field. Parameters are given in the text.

in terms of the Floquet theory, assuming classical behavior of this oscillatory-in-time field, whereas the interaction with the waveguide mode reflects a quantum interference. The obtained results demonstrate the high potential of waveguide polaritons for all-optical switching. Both, frequency and amplitude of the external laser control transmission through the waveguide, and each of these features ensure ultrafast switching processes.

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